First conclusions of the WPEC/Subgroup-22 Nuclear Data For Improved LEU-LWR Reactivity Predictions DRAFT COPY FOR DISCUSSION

WPEC/subgroup-22 *www.nea.fr/lists/ueval*
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1 Main contributions

This paper is a summary of a co-operative work compiled in the framework of the WPEC/Subgroup-22. A more detailed report will follow. The main contributors are listed below, the coordinator apologises persons whose names and work are not explicited cited in this report.

- **International Atomic Energy Agency (IAEA)** :
- **Bettis Atomic Power Laboratory (BAPL)** :
- **CEA-Cadarache** :
- **CEA-Bruyeres le Chatel (CEA-BRC)** :
- **Japan Atomic Energy Research Institute (JAERI)** :
- **Knoll Atomic Power Laboratory (KAPL)** :
- **Los Alamos National Laboratory (LANL)** :
- **Nuclear Research and Consultancy Group (NRG) Petten** :
- **Oak Ridge National Laboratory (ORNL)** :
- **Serco Assurance Winfrith** :
- **Westinghouse Electric Corp** :
- **other**

2 Introduction

The subgroup-22 was created at the WPEC Meeting, held at GEEL 23-24 May 2001, to address the so-called "reactivity under-prediction problem" for thermal LEU-LWR (**L**ow-**E**nriched-**U**ranium, **L**ight **W**ater **R**eactor) systems. The communication within the group is mainly assured through the NEA mailing list called **ueval** (uranium evaluation) *ueval@nea.fr*. The initial subgroup proposal is available at the WPEC web page *www.nea.fr/html/science/wpec/index.html* and at *http://www.nea.fr/lists/ueval.html*.

3 Trends derived from integral experiments

3.1 Under-estimation of thermal LEU-LWR k_{eff}

Before the creation of the WPEC working group, several Continuous Energy Monte-Carlo (CEMC) studies had raised the problem [1], [2], [3], [4]. The reactivity bias was observed for low enriched thermal benchmarks after extensive integral tests of the ORNL U235 evaluation of the resolved resonances by Leal, Derrien, Wright and Larson [5] adopted in ENDF/B-VI.8, JEFF3.0 and JENDL3.3. A summary of the work done on U235 can be found in the final report of the WPEC/subroup-18 [6]. It was demonstrated that the ORNL U235 evaluation improves k_{eff} of Highly Enriched Uranium critical systems and corrects the longstanding underestimation of U236 build-up in PWR. However, recent studies by Kahler et al. [7], Weinman et al. [8] and Van Der Marck and Hogenbirk [2] involving low enriched thermal benchmarks mainly from the ICSBEP and CSEWG benchmarks book confirmed a systematic eigenvalue under-estimation of about -400 pcm (depending slightly on the library used) with the newest nuclear data libraries : ENDFB/VI.8, JEFF3.0 and JENDL3.3.

3.2 U238 integral trends in thermal and epithermal range

The resonance parameters of U238 are the same in ENDF/B-VI.8, JEFF3.0 and JENDL3.3 and are the result of an evaluation work performed by Moxon et al. [9]. Integral trends on U238 capture cross-section have been obtained by means of k_{eff} bias analysis as a function of U238 capture fraction (Kahler [7] and Weinman [8]), interpretation of U238 spectral indices : ρ^{28}, $C^* = \sigma_c^{U238}/\sigma_f^{U235}$, C^{U238}/F^{tot} performed in the EOLE facility in France [10], in the IPEN brazilian mock-up [11] and in TRX and DIMPLE reactor [12], analyses of Hellstrand experimental correlations [13] [14] (measurements of effective capture resonance integral as a function of $\sqrt{S/M}$ [15], [17], [16]) and Post-Irradiation Experiments (prediction of Pu239 production versus burn-up) [18].

The conclusion drawn from those studies is that no significant discrepancies are so far demonstrated with the U238 evaluation of Moxon : the C/E values are generally within the uncertainty of the integral measurements demonstrating the quality of the present resonance parameters set. The results are nevertheless compatible with a small decrease of the U238 effective capture resonance integral in the resonance range by about 0.5% to 1.0% :

4 Evaluation work

4.1 U238 thermal capture cross-section value

Different recommended values for the U238 thermal capture cross-section can be found : $\sigma_0 = 2.708\ b$ in the CSEWG recommendation [19], $\sigma_0 = 2.718\ b$ with the positive and negative resonance parameters of Moxon and 2.680 ± 0.019 b from the latest Mughabghab review of integral quantities [20] [21]. The U238(n,γ) thermal cross-section was recently reviewed by A. Trkov et al. [23]. Some of the activation data selected for the evaluation are based on the measurements of gamma activity from the beta decay of Np239. Because the derived cross-section value is strongly correlated to the knowledge of gamma-ray emission probability, a review of exisiting measurements of gamma line production data was also performed. In addition, the measurements carried out in Budapest and published recently [24] were reanalysed in depth to get new gamma ray experimental data. The final least squares fit was performed with the ZOTT99 code in the log-domain to avoid the so-called "Peele Pertinant Puzzle" widely discussed in the field of standards cross-section evaluation and the final value is : $\sigma_0 = 2.683 \pm 0.012\ b$ New evaluations of several Np239 gamma-ray emission probabilities are also proposed in this study submitted to Nuclear Science and Engineering.

4.2 U238 resolved resonance range

resonance parameters of Moxon An extensive work through a NEANDC task force on U238, coordinated by M. Moxon and M. Sowerby, led in 1990 to an improved evaluation of the U238 resonance range up to 10 keV which was included in the most recent libraries except BROND2. The work done at that time on U238 is summarised in [9] and in the final report of the WPEC/Subgroup-4 "U238 capture and inelastic cross-sections" [25]. Selected capture and transmission measurements, listed in reference [9] were simultaneous fitted by M. Moxon using the Reich-Moore formalism of the shape analysis code REFIT with improved modelling of resolution functions. The main conclusions reached by the U238 task force explained the major discrepancies between differential measurements and the set of resonance parameters from Moxon was expected to solved the longstanding overestimation of the U238 capture cross-section derived from reactor analyses (see the proceedings of the 1975 U238 seminar at Brookhaven for a description of the problems encountered [26]).

Adjusted cross-section for sensitivity studies In the light of the integral trends suggesting a small reduction of the capture cross-section in the resolved range, Cecil Lubitz (KAPL) proposed two different adjustments of the U238 resonance parameters and distributed ENDF formatted files to the working group for sensitivity studies. The first adjustment (KAPL-22-1) is mainly based on a uniform reduction by 1.35% average radiation widths of the reduced s-wave positive-energy whereas the second (KAPL-22-2) featured neutron widths lowered by 0.738%.

The dilute capture resonance integral (276.6 b from 0.5 eV to 20 MeV) and thermal capture cross-section (2.708 b) have the same values in the two adjustments. However, the effect of these changes on effective capture resonance integral (with dilution values in the range 20 - 100 barns) was found to be not strictly equivalent [29].

Status of the ORNL work on U238 resonance parameters A new evaluation of resonance parameters is in progress at the Oak Ridge National Laboratory (H. Derrien, L. Leal, N. Larson). The status of the evaluation was presented at the PHYSOR-2004 conference [30]. Sequential fits of differential measurements listed in Table 1 were performed with the Reich-Moore formalism implemented in SAMMY. Note that the Harvey et al. high resolution measurements [33] performed at ORELA in 1988 were used for the first time in the U238 evaluation extending the resolved range from 10 keV to 20 keV. The important capture data of Macklin [34] (ORELA) from 1 keV to several hundred keV and other capture data from Harwell used [35] in the Moxon's evaluation were not available for this preliminary evaluation.

932 s-wave and 2354 p-wave resonances have been identified. The effective radius, obtained from fits of transmissions between resonances, is 9.450 fm close to the Olsen [31] ($9.44 \pm 0.005 fm$) and Moxon analysis (9.428 fm). Negative energy resonances were slighly adjusted to have the thermal capture cross-section close to $\sigma_0 = 2.680\ b$. Only the neutron widths have been adjusted (radiation widths were kept fixed in the present fit to the moxon's values). The neutron widths of the first three resonances 6.7 eV, 20.8 eV and 36.7 eV are slighly smaller than the Moxon's

Authors	Energy range	Number of samples	exp. methods
Olsen et al. [31]	5 eV - 10 keV	7	transmission
Olsen et al. [32]	300 eV - 20 keV	4	transmission
Harvey et al. [33]	1 keV - 20 keV	3	transmission
De Saussure al. [36]	6 eV - 900 eV	2	capture

Table 1: Differential Measurements used for the preliminary resolved range evaluation of ORNL

leading to a smaller effective capture resonance integral still compatible with U238 integral trends derived in Section 2.

Influence of solid state effects on U238 resonance parameters For the broadening of the low energy resonances of U238, the current evaluations (Moxon, ORNL) are based on fits using the Free Gas Model (FGM) with fitted temperature (which vary from resonance to resonance) to account for solid state effect. It was demonstrated in [37] by D. Nabarejnev et al. using the Crystal Lattice Model (CLM) [38] implemented in the DOPUSH program, that FGM with fitted temperature could, in certain cases produce significant bias on extracted resonance parameters. The DOPUSH code, was recently put into SAMMY and REFIT with some minor modifications. The CLM of SAMMY code was tested [40]) against transmission measurements performed at GEEL [39] (uranium oxyde and metallic sample measured at 23.7K and room temperature. The strong assymetry of the resonance shape, produced by crystalline binding, is well reproduced by the CLM of SAMMY using appropriate phonon spectrum. It is expected to include CLM model in the final ORNL U238 fit.

4.3 U238 inelastic scattering data

In the past 20 years, a great effort has been devoted to the improvement of the modeling of U238 cross-section above the unresolved range. In 1989, under the framework of WPEC/Subgroup-4 [25] a set of measurements on U238(n,n') were performed to solve discrepancies and reduce uncertainties in the previous data. Experimental neutron spectrum, partial cross-section measurements listed in [25] gave valuable information on optical model parameters. In the meantime, coupled channel and statistical models have been improved to predict U238 inelastic cross-section. As a result of subgroup-4 efforts, improved evaluations of U238 have been released by Maslov et al. [42] and Kawano et al. [43]. Recently, independant evaluations by the Los-Alamos [44] and Bruyeres Le Chatel [45] group were undertaken and successive releases distributed to the subgroup-22 for discussion and testing purpose. Compared to older ENDFB/VI.8 and JEF2.2 evaluation, performed before the set-up of the subgroup-4, the comparison with experimental data, in particular, neutron emission spectra (inelasticaly scattered neutron + fission) at different incident energy, is much improved by these new files. The influence of these new data on ICSBEP thermal benchmarks was found to correct partially the reactivity bias by reducing the neutron leakage rate. A sensitivity study have also been performed by C. Lubitz and A. Kahler to assess the effect of representing inelastic-scattering levels as a continuum in reactor calculations. The replacment of continuum data by pseudo-levels was found to have a small effect in benchmark calculation.

5 Status of Integral testing

The different versions of evaluations in the resolved range (KAPL, ORNL) and above the unresolved range (BRC, LANL), have been continuously tested against k_{eff} measurements. Some intercomparisons have demonstrated the overall consistency of the calculated eigenvalues between Monte-Carlo codes (MCNP - RACER - RCP - TRIPOLI4) using the same nuclear data libraries. Some differences were sometimes noticed (up to 150 pcm for some leu-Comp-Therm configurations between TRIPOLI4 and MCNP) and, so far, could not be explained by the statistical uncertainty inherent to Monte-Carlo calculations. Investigation of these differences would require a more rigorous comparison (neutron balance, reaction rate and so on.) to identify whether it comes from differences in benchmarks modelling, processing methodology, transport methods or assessment of statistical uncertainty in Monte-Carlo codes.

In the resolved range, the two KAPL adjustments and ORNL preliminary evaluation featured a lower effective capture resonance integral than the Moxon's evaluation : about -0.2% (KAPL-22-2), -0.4% (ORNL) to -0.7% (KAPL-22-1) in the dilution range 20 - 100 barns and a lower thermal capture value. The effect is an expected increase of the calculated multiplication factor for low-enriched lattices from about 100 to 200 pcm depending of the moderation ratio.

Compared with the data in JEF2.2 and ENDF/B-VI.8, the latest versions of inelastic scattering data from LANL and BRC also produced a similar increase of the k_{eff} for low-enriched thermal lattices with high neutron leakage rate mainly because of a softer secondary inelastic energy spectrum in the fission range.

The preliminary resolved (ORNL) and fast range data (LANL, BRC) have been eventually merged in two complete U238 test files (ORNL + LANL) and (ORNL + BRC). Integral testing of these files has started but the expected improvement of reactivity prediction for Low-enriched thermal lattices of ICSBEP was confirmed by the first studies ([46], [47] for JEFF3.0 + U238 ORNL-BRC and [48] for ENDF/B-VI.8 + U238 ORNL-LANL results). It was also checked that the present new evaluation of U238 data preserves a good C/E agreement (with JEFF3.0 and ENDF/B-VI.8) for other kind of uranium systems such as Leu-Sol-Therm, Heu-Sol-Therm. In addition, with the latest versions of U238 files, fast benchmarks are found to be fairly well predicted [49].

6 Other nuclear data impacting LEU-LWR k_{eff}

O16 : Large differences between modern libraries are observed for the O16 (n, α) cross-section above 3 MeV. In JENDL3.2 O16 (n, α) is significantly lower than in ENDF/B-VI.8 and JEF2.2. In the recent analysis of Hale et al. [50] and Sayer et al. [51] $\sigma_{n,\alpha}$ cross-sections are generally deduced by reciprocity from $C13(\alpha, n)$ measurements. However, in the energy range 3-6 MeV, large differences (10% to 50%) are noticed between the data [51] and a new measurement would help to remove those discrepancies.

U235 prompt fission spectrum : At the 2003 WPEC Meeting in San-Diego, David Madland presented the work of sub-group-9 on the evaluation of fission neutron spectra. In the final report [52], it is pointed out large discrepancies in the peak and in the tail region between the two most recent measurements of U235 spectrum for thermal neutron. The latest evaluation proposed by David Madland is still preliminary but the current uncertainty on the shape of U235 thermal spectrum has a strong impact on k_{eff} of HEU and LEU criticality systems. Despite the work done at Los Alamos to investigate the discrepancy and improve theoretical modelling, a highly accurate measurement of the U235 thermal fission spectrum is strongly needed

7 First conclusions

During the three years of the subgroup-22, the preliminary conclusions have been drawn :

- The underestimation of k_{eff} of about 400 pcm of Low-Enriched Uranium lattices is confirmed with ENDFB/VI.8, JEFF3.0 and JENDL3.2 and is not likely the consequence of numerical approximations in reactor calculation methods. This point was checked using Monte-Carlo approaches and accurate pointwise representation of nuclear data.

- Given the large number of independant low enriched experiments investigated (mostly from the ICSBEP handbook), the present reactivity bias is not believed to come from experimental errors in criticality measurements.

- The problem has been studied on small size configurations (high neutron leakage rate) and so far, is not demonstrated (with CEMC code) for large commercial PWR.

- Given the improvements brought by the ORNL evaluation of U235 resonance range (prediction of Highly Enriched Uranium systems and U236 isotopic ratios in Post-Irradiation Experiments), this evaluation is probably not the source of the underestimation.

- Present integral tests of the current U238 resonance parameters from Moxon et al. adopted in ENDFB/VI.8, JEFF3.0 and JENDL3.3 do not demonstrate a significant bias. Integral experiments (spectral indices, Pu239 production in PWR) are predicted within the experimental uncertainty margins. A slight reduction of effective resonance integral between 0.5% to 1% is nevertheless compatible with integral experiments and would improve thermal lattices k_{eff} prediction.

- A new evaluation of the thermal (2200 m/s) U238 capture cross-section is proposed : $\sigma_0 = 2.683 \pm 0.012\ b$.

- The preliminary evaluation of ORNL from 0 to 20 keV which features a lower thermal capture cross-section and effective resonance integral compared with the evaluation of Moxon (in the dilution range 20 - 100 barns) corrects partly the k_{eff} underestimation.

- The new evaluations of U238 inelastic scattering data from LANL and BRC also increase ICSBEP Leu-Comp-Therm k_{eff} and contribute to the correction of the underestimation.

- The combination of the new inelastic data (LANL or BRC) with the preliminary ORNL resonance parameters set gives a satisfactory correction of the reactivity under-estimation.

- New differential measurements are recommended :

  - A measurement of the U238 (n,γ) in the resolved range is recommended (ideally from the thermal range to 1 keV). The priority is to measure the U238 (n,γ) cross-section below 120 eV with the highest accuracy achievable.
  - In the energy range 3-6 MeV, large differences (10% to 50%) are noticed between the evaluations of O16 (n, α) and a new measurement would help to remove these discrepancies.
  - A measurement of U235 prompt fission spectrum for incident neutron at thermal energy is desirable as recommended by the WPEC/Subgroup-9.

8 Proposal for extension of the working group duration

The present results are based on an interim version of resonance parameters file from ORNL. Work is still in progress at ORNL to improve it and several actions are underway : inclusion of the capture measurements of Macklin et al. in the fit, use of Crystal Lattice Model implemented in SAMMY for the broadening of the low energy resonances, evaluation of U238 unresolved range. The Los-Alamos group also plan to perform additional modifications of their uranium evaluations above the unresolved resonance range.

Moreover, the complete U238 files including latest improvements (ORNL-LANL and ORNL-BRC) have been tested against k_{eff} measurements but further tests would be beneficial : k_{eff} of critical systems reflected by depleted uranium, spectral indices and effective resonance integral measurements, production of Pu239 in Post-Irradiation Experiments, thick samples Transmission and self-indication measurements for the validation of unresolved range (see the measurements [53], [54]), pulsed sphere experiments with neutron leakage spectra measurements (see for instance hansen et al. [55]).

Given the work in progress to improve and test existing evaluations, a 1 year extension of the subgroup duration is proposed to strengthen the present conclusions.

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